

FORM PTO-1390 (REV. 9-2001)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE <b>TRANSMITTAL LETTER TO THE UNITED STATES          DESIGNATED/ELECTED OFFICE (DO/EO/US)          CONCERNING A FILING UNDER 35 U.S.C. 371</b>		ATTORNEY'S DOCKET NUMBER <b>566.41199X00 filed February 21, 2002</b>	
INTERNATIONAL APPLICATION NO. <b>PCT/JP00/05763</b>		INTERNATIONAL FILING DATE <b>August 25, 1999</b>		U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <b>10/069053</b>	
TITLE OF INVENTION <b>WIRING-CONNECTED MATERIAL AND WIRING-CONNECTED BOARD PRODUCTION PROCESS USING THE SAME</b>					
APPLICANT(S) FOR DO/EO/US <b>FUJINAWA, TOHRU YUSA, MASAMI NOMURA, SATOYUKI ONO, HIROSHI KANAZAWA, HOUKO</b>					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.					
2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.					
3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.					
4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).					
5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))					
a. <input type="checkbox"/> is transmitted hereto (required only if not communicated by the International Bureau).					
b. <input checked="" type="checkbox"/> has been communicated by the International Bureau.					
c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office(RO/US)					
6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).					
a. <input checked="" type="checkbox"/> is attached hereto.					
b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).					
7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))					
a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).					
b. <input type="checkbox"/> have been communicated by the International Bureau.					
c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.					
d. <input type="checkbox"/> have not been made and will not be made.					
8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).					
9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).					
10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).					
<b>Items 11 to 20 below concern document(s) or information included:</b>					
11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.					
12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.					
13. <input type="checkbox"/> A FIRST preliminary amendment.					
14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.					
15. <input type="checkbox"/> A substitute specification.					
16. <input checked="" type="checkbox"/> A change of power of attorney and/or address letter.					
17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.					
18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).					
19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).					
20. <input checked="" type="checkbox"/> Other items or information: <b>Figs. 1a-1d, 2-3, Credit Card Payment Form, International Search Report, International Preliminary Examination Report, International Publication No. WO 01/15505-coversheet</b>					

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)		INTERNATIONAL APPLICATION NO.		ATTORNEY'S DOCKET NUMBER	
10/069053		PCT/JP00/05763		566.41199X00	
21. The following fees are submitted <b>BASIC NATIONAL FEE (37 CFR 1.492(a) (1) - (5)):</b> <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO . . . . . \$1040.00  <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO . . . . . \$890.00  <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. . . . . \$740.00  <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4). . . . . \$710.00  <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) . . . . . \$100.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total Claims	13 - 20 =	0	x \$18.00	\$	
Independent Claims	2 - 3 =	0	x \$84.00	\$	
MULTIPLE DEPENDENT CLAIMS(S) (if applicable) 280.00			+ \$280.00	\$280.00	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$1,170.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2				\$	
<b>SUBTOTAL =</b>				\$1,170.00	
Processing fee of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f))				\$	
<b>TOTAL NATIONAL FEE =</b>				\$1,170.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	
<b>TOTAL FEES ENCLOSED =</b>				\$1,170.00	
				Amount to be refunded:	\$
				charged:	\$
a. <input type="checkbox"/> A check in the amount of \$_____ to cover the fees is enclosed  b. <input type="checkbox"/> Please charge my Deposit Account No. <b>01-2135</b> in the amount of \$_____ to cover the above fees A duplicate copy of this sheet is enclosed  c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposition Account No. <b>01-2135</b> . A duplicate copy of this sheet is enclosed  d. <input checked="" type="checkbox"/> Fees are to be charged to a credit card. <b>WARNING:</b> Information on this form may become public. <b>Credit card information should not be included on this form.</b> Provide credit card information and authorization on PTO-2038.					
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>					
SEND ALL CORRESPONDENCE TO  <b>Antonelli, Terry, Stout &amp; Kraus, LLP</b> 1300 North Seventeenth Street Suite 1800 Arlington, VA 22209 USA					
				 SIGNATURE	
				<b>Alan E. Schiavelli</b> NAME	
				<b>32,087</b> REGISTRATION NO.	

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566.41199X00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



Applicant(s): FUJINAWA, et al.

Serial No.: 10/069,053

Filed: February 21, 2002

For: WIRING-CONNECTED MATERIAL AND WIRING-  
CONNECTED BOARD PRODUCTION PROCESS USING  
THE SAME

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231

May 21, 2002

Sir:

Please amend the above-identified application, prior to examination  
thereof, as follows:

IN THE CLAIMS

Please amend the claims presently in the application as follows:

6. (Amended) The wiring-connecting material according to any one  
of claims 1 to 3, wherein said curing agent has a weight retention at 25°C for 24  
hours, of not less than 20% by weight.

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--10. The wiring-connecting material according to claim 4, wherein said curing agent has a weight retention at 25°C for 24 hours, of not less than 20% by weight.

11. The wiring-connecting material according to claim 5, wherein said curing agent has a weight retention at 25°C for 24 hours, of not less than 20% by weight.

12. The wiring-connecting material according to claim 4, wherein said radical-polymerizable substance is urethane acrylate.

13. The wiring-connecting material according to claim 5, wherein said radical-polymerizable substance is urethane acrylate.

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radical-polymerizable substance is urethane acrylate.--



filing of this paper, including extension of time fees, to the Deposit Account No. 01-2135 (Case No. 566.41199X00) and please credit any excess fees to such Deposit Account.

Respectfully submitted,

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WIS/slk

VERSION WITH MARKINGS TO SHOW CHANGES MADE

Please amend the claims presently in the application as follows:

6. (Amended) The wiring-connecting material according to any one of claims 1 to [5] 3, wherein said curing agent has a weight retention at 25°C for 24 hours, of not less than 20% by weight.

7. (Amended) The wiring-connecting material according to any one of claims 1 to [6] 3, wherein said radical-polymerizable substance is urethane acrylate.



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## DISCRIPTION

### WIRING-CONNECTING MATERIAL

### AND

### WIRING-CONNECTED BOARD PRODUCTION PROCESS USING THE SAME

## TECHNICAL FIELD

This invention relates to a wiring-connecting material making use of an adhesive composition and conductive particles, and a process for producing a wiring-connected board.

5

## BACKGROUND ART

Epoxy resin adhesives are widely used for various purposes such as electric, electronic, construction, automobile and airplane materials because they can provide a high bond strength and have excellent resistance to water and resistance to heat.

10

In particular, one-pack type epoxy resin adhesives are usable with ease because it is unnecessary to blend the chief material and the curing agent, and are widely used in the form of film, paste or powder. In such one-pack type epoxy resin adhesive, epoxy resins, curing agents and modifiers can be combined in variety. Hence, appropriate selection of their combination enables achievement of any desired performance as disclosed in, e.g., Japanese Patent Application Laid-open (KOKAI) No. Sho62-141083.

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A filmlike adhesive disclosed in this Japanese Patent Application Laid-open (KOKAI) No. Sho62-141083, however, can not sufficiently be reactive at the time of curing because it uses a catalytic curing agent which is inert at normal temperature, 5 in order to achieve both short-time curing properties (fast-curing performance) and storage stability (storage properties) to attain a good stability. Thus, though having a good operability, it must be heated at about 140°C to about 180°C for a connecting time of about 20 seconds, or must be 10 heated at about 180°C to about 210°C for a connecting time of 10 seconds.

In recent years, however, in the field of precision electronic instruments, circuits are being made highly dense, so that connecting terminals have come to be formed in and at 15 very narrow width and pitch. Hence, this has caused come-off or peeling and misregistration of wiring in some cases when terminals are interconnected under connecting conditions adapted to such conventional epoxy resin wiring-connecting materials.

Also, it has been sought to shorten connecting time so as to 20 enable connection within 10 seconds in order to improve production efficiency. To satisfy these demands, it is sought to provide a low-temperature fast-curable wiring-connecting material capable of curing at a low temperature and yet in a short time.

An object of the present invention is to provide a wiring-connecting material for electric and electronic use, having better low-temperature fast-curing performance than any conventional epoxy resin connecting materials and also having a pot life, and a process for producing a wiring-connected board making use of such a connecting material.

The wiring-connecting material of the present invention may also preferably further comprise a film-forming material and/or conductive particles. The film-forming material may preferably be mixed in an amount of from 0 to 40 parts by weight, and a polyimide resin is preferred.

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to use a curing agent having a weight retention at 25°C for 24 hours (i.e., the proportion of a difference in mass before and after open-leaving at room temperature (25°C) and normal pressure for 24 hours, with respect to the mass before leaving)  
 5 of not less than 20% by weight. As the radical-polymerizable substance, urethane acrylate is preferred.

The present invention also provides a wiring-connected board production process comprising the step of interconnecting terminals of wiring by the use of the wiring-connecting material  
 10 of the present invention. More specifically, the present invention provides a process for producing a wiring-connected board, comprising a connecting step of interconnecting wiring members each having a connecting terminal, which members are so interconnected that their connecting terminals are able to  
 15 make conduction between them;

the connecting step comprising the step of heating the wiring-connecting material of the present invention while applying a pressure thereto via the wiring members; the wiring-connecting material being held between at least two wiring  
 20 members so disposed that their sides having the connecting terminals face to each other. This production process of the present invention is especially preferable when at least one surface of the connecting terminal is formed of at least one metal selected from gold, silver and a platinum group metal.

25

Figs. 1(a) to 1(d) illustrate a wiring-connected board production process carried out in Example 1.

Fig. 3 is a cross-sectional view showing connecting portions on a wiring substrate of a liquid-crystal display panel, used in Example 4.

### A. Curing Agent

The curing agent used in the present invention is a curing agent capable of generating a free radical upon decomposition by heating, such as a peroxide compound or an azo compound. The curing agent may appropriately be selected in accordance with the intended connecting temperature, connecting time, pot life and so forth. In order to achieve a high reactivity and a long pot life, preferred is an organic peroxide showing a 10-hour half-life at a temperature of 40°C or above and a 1-minute half-life at a temperature of 180°C or below, and more preferred is an organic peroxide showing a 10-hour half-life at a temperature of 60°C or above and a 1-minute half-life at a temperature of 170°C or below.

When the connecting time is made not longer than 10  
25 seconds, the curing agent may preferably be mixed in an amount

10           The organic peroxide preferred as the curing agent used in the present invention is exemplified by diacyl peroxides, peroxydicarbonates, peroxyesters, peroxyketals, dialkyl peroxides, hydroperoxides and silyl peroxides.

The diacyl peroxides may include isobutyl peroxide,  
15 2,4-dichlorobenzoyl peroxide, 3,5,5-trimethylhexanoyl  
peroxide, octanoylperoxide, lauroylperoxide, stearoylperoxide,  
succinic peroxide, benzoyl peroxytoluene and benzoyl peroxide.

The peroxydicarbonates may include di-n-propyl peroxydicarbonate, diisopropyl peroxydicarbonate, bis(4-t-butylcyclohexyl) peroxydicarbonate, di-2-ethoxyethyl peroxydicarbonate, di(2-ethylhexyl peroxy)dicarbonate, dimethoxybutyl peroxydicarbonate, and di(3-methyl-3-methoxybutyl peroxy)dicarbonate.

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peroxyneodecanoate, t-butyl peroxyphosphate,  
1,1,3,3-tetramethylbutyl peroxy-2-ethylhexanone,  
2,5-dimethyl-2,5-di(2-ethylhexanoyl peroxy)hexane,  
1-cyclohexyl-1-methylethyl peroxy-2-ethylhexanone, t-hexyl  
5 peroxy-2-ethylhexanone, t-butyl peroxy-2-ethylhexanone,  
t-butyl peroxyisobutyrate, 1,1-bis(t-butylperoxy)cyclohexane,  
t-hexyl peroxyisopropylmonocarbonate, t-butyl  
peroxy-3,5,5-trimethylhexanone, t-butyl peroxy laurate,  
2,5-dimethyl-2,5-di(m-toluoylethoxy)hexane, t-butyl  
10 peroxyisopropylmonocarbonate, t-butyl  
peroxy-2-ethylhexylmonocarbonate, t-hexyl peroxybenzoate, and  
t-butyl peroxyacetate.

The dialkyl peroxides may include

The hydroperoxides may include diisopropylbenzene hydroperoxide and cumene hydroperoxide.

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t-butyltrivinylsilyl peroxide, bis(t-butyl)divinylsilyl peroxide, tris(t-butyl)vinylsilyl peroxide, t-butyltriallylsilyl peroxide, bis(t-butyl)diallylsilyl peroxide, and tris(t-butyl)allylsilyl peroxide.

5           In the present invention, as the curing agent, any one of these compounds may be used or any two or more compounds may be used in combination. Also, any of these curing agents (free-radical generators) may be used in combination with a decomposition accelerator, an inhibitor and so forth.

10           In order to keep the connecting terminals of wiring members from corroding, chloride ions and organic acids contained in the curing agent may preferably be in a content not more than 5,000 ppm. Those generating less organic acid after heat decomposition are more preferred. Also, on account of an  
15   improvement in the stability of wiring-connecting materials produced, the curing agent may preferably have a weight retention of not less than 20% by weight after open-leaving at room temperature (25°C) and normal pressure for 24 hours.

          Any of these curing agents may also be coated with,  
20   e.g., a polymeric material of polyurethane type or polyester type so as to be made into microcapsules. Such a curing agent is preferred because of a prolonged pot life.

#### B. Polyurethane Resin

          As the polyurethane resin, a resin obtained by the reaction  
25   of a diol, having two hydroxyl groups in the molecule, with a diisocyanate, having two isocyanate groups in the molecule,



As the diol, any of those which are linear compounds  
5 and having hydroxyl groups at the terminals may be used. Stated  
specifically, it may include polyethylene adipate,  
polydiethylene adipate, polypropylene adipate, polybutylene  
adipate, polyhexamethylene adipate, polyneopentyl adipate,  
polycaprolactone polyol, poly(hexamethylene carbonate),  
10 silicone polyol, acrylic polyol, poly(ethylene glycol),  
poly(propylene glycol) and poly(tetramethylene glycol). Any  
of these compounds may be used alone, or may be used in combination  
of two or more types.

The diisocyanate may include isophorone diisocyanate, tolylene diisocyanate, 4,4'-diphenylmethane diisocyanate, naphthalene-1,5-diisocyanate, p-phenylene diisocyanate, 4,4'-methylenebiscyclohexyl diisocyanate, hexamethylene diisocyanate, and cyclohexane diisocyanate. Any of these compounds may be used alone, or may be used in combination of two or more types.

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The film-forming material used in the present invention may include polyimide resins, polyvinyl formal resins, polystyrene resins, polyvinyl butyral resins, polyester resins, acrylic resins, polyamide resins, xylene resins and phenoxy resins. Any of these may be used alone, or may be used in combination of two or more types.

The film-forming material is a material providing mechanical properties that enable films to be handled as films in an ordinary condition, such that, when a liquid material is made into a solid and its composition made up is formed into a self-supporting film, the resultant film can be handled with ease and not be readily broken or cracked or sticky.

As the film-forming material, polyimide resin is especially suited for the present invention in view of resistance to heat. Usable as the polyimide resin are, e.g., those obtained by subjecting a polyamic acid synthesized by addition reaction of a tetracarboxylic dianhydride with a diamine to heating and condensation to effect imidization. The polyimide resin may preferably have a weight-average molecular weight of from about 10,000 to about 150,000 in view of film-forming properties.

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Methacrylates corresponding to these acrylates may also be preferable in the present invention.

Radical-polymerizable substances having at least one of a dicyclopentanyl group, a tricyclodecanyl group and a triazine ring are preferred because the wiring-connecting material obtained can be improved in resistance to heat.

As the radical-polymerizable substance used in the present invention, urethane acrylates are particularly preferred because of their superior adhesion. The urethane acrylates are those having at least one urethane group in the molecule, and may include, e.g., reaction products of polyols such as poly(tetramethylene glycol) with polyisocyanates and hydroxyl-group-containing acrylic compounds.

On account of an improvement in bond strength on the surfaces of inorganic matters such as metals, a radical-polymerizable substance further having a phosphate structure may also preferably be used in combination, in addition to the radical-polymerizable substance described above.

Such a radical-polymerizable substance having a phosphate structure, preferable in the present invention, may include reaction products of phosphoric anhydride with 2-hydroxyethyl acrylate or its corresponding methacrylate 2-hydroxyethyl methacrylate. Stated specifically, mono(2-methacryloyloxyethyl)acid phosphate and di(2-methacryloyloxyethyl)acid phosphate may be used. Any of these compounds may be used alone, or may be used in combination

As the maleimide compounds, those containing at least two maleimide groups in the molecule are suited for the present invention. Such maleimide compounds may include, e.g.,

In the present invention, in addition to these radical-polymerizable substances, polymerization inhibitors such as hydroquinones and methyl ether hydroquinones may optionally appropriately be used.

In the wiring-connecting material of the present

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invention, the above respective ingredients may be contained in an amount of from 2 to 75 parts by weight for the polyurethane resin, from 30 to 60 parts by weight for the radical-polymerizable substance, and from 0.1 to 30 parts by weight for the curing agent capable of generating a free radical upon heating. In the case when the film-forming material is contained, it may be contained in an amount of from 0 to 40 parts by weight. These ingredients may be mixed in the amounts appropriately determined within the above ranges.

10           If the polyurethane resin is in a content smaller than 2 parts by weight, the wiring-connecting material may have a poor effect of stress relaxation at the time of heat loading to lower bond strength. If it is in a content larger than 75 parts by weight, there is a possibility of a low connection  
15   reliability.

          If the radical-polymerizable substance is in a content smaller than 30 parts by weight, the wiring-connecting material tends to have a low mechanical strength after its curing. If it is in a content larger than 60 parts by weight, the  
20   wiring-connecting material may have a high tackiness before its curing, resulting in poor handling properties.

          If the curing agent capable of generating a free radical upon heating is in a content smaller than 0.1 part by weight, no sufficient rate of reaction can be attained as stated previously,  
25   to tend to make it difficult to attain a good bond strength and a small connection resistance. Also, if this curing agent

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is in a content larger than 30 parts by weight, the wiring-connecting material tends to have low flow properties, cause a high connection resistance and have a short pot life.

If the film-forming material is in a content more than  
5 40 parts by weight, the wiring-connecting material tends to have low flow properties and cause a high connection resistance.

Incidentally, the film-forming material need not be contained as long as other ingredients such as the polyurethane resin, the radical-polymerizable substance and the curing agent capable  
10 of generating a free radical upon heating enable sufficient film formation.

#### F. Conductive Particles

The wiring-connecting material of the present invention can interconnect the connecting terminals through their direct  
15 contact even when it does not contain any conductive particles.

Hence, it need not particularly contain any conductive particles.

However, on account of achievement of stabler connection, it may preferably contain conductive particles.

As the conductive particles, usable are particles of  
20 metals such as Au, Ag, Ni, Cu and solder, carbon particles or the like. In order to attain a sufficient pot life, the surface layers of particles may preferably be formed of not a transition metal such as Ni or Cu but a noble metal such as Au, Ag or a platinum group metal, and particularly preferably be formed  
25 of Au.

Composite particles comprised of a transition metal



Coat layers of a noble metal in such composite particles may be formed in a thickness of, but not particularly limited to, 100 Å or larger in order to achieve a good resistance. When, however, the layers of a noble metal are provided on the surfaces of a transition metal such as Ni, noble-metal layer defects caused when the conductive particles are mixed and dispersed, may cause the action of oxidation-reduction to generate free radicals to make the pot life short. Accordingly, the noble-metal layers may preferably be in a thickness 300 Å or larger. Also, if the noble-metal layers have a thickness larger than 1 μm, the effect may become saturated, and hence a layer thickness not larger than 1 μm is effective in view of cost and so forth.

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terminals, bringing about an improvement in reliability.

In the case when the conductive particles are mixed in the wiring-connecting material of the present invention, they may be mixed in an amount of from 0.1 to 30% by volume based on the adhesive component, which may appropriately be determined depending on purposes. In order to prevent adjoining circuits from short-circuiting due to any excess conductive particles, they may preferably be mixed in an amount of from 0.1 to 10% by volume.

#### 10 G. Additives

In addition to the ingredients A to F described above, the wiring-connecting material of the present invention may further contain a filler, a softener, an accelerator, an anti-aging agent, a colorant, a flame retardant, a thixotropic agent, a coupling agent and so forth.

The compounding of a filler is preferred because the wiring-connecting material obtained can be improved in connection reliability and so forth. The filler may preferably have a maximum diameter that is smaller than the particle diameter of the conductive particles, and it may be compounded in an amount of from 5 to 60% by volume. If it is in an amount more than 60% by volume, the effect of improving reliability is saturated.

As the coupling agent, agents containing a vinyl group, an acrylic group, an amino group, an epoxy group or an isocyanate group are preferred in view of an improvement in adhesion.

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## H. Film Structure

The wiring-connecting material constituted as described herein need not be in the form of a single layer in which all the ingredients are present, and may be in the form of a stacked  
 5 film having two or more layers. For example, it may have a double-layer structure consisting of a layer containing the curing agent capable of generating a free radical upon heating and a layer containing the conductive particles to separate these ingredients. This can bring about not only the effect  
 10 of enabling achievement of high precision but also the effect of improving pot life.

## I. Properties of Wiring-connecting Material

The wiring-connecting material of the present invention interconnects the connecting terminals facing to each other,  
 15 by bringing them into contact with the adhesive which melt-flows at the time of connection, followed by curing to keep their connection, and thus the flow properties of the adhesive is an important factor. The wiring-connecting material of the present invention may preferably have a value of flow properties  
 20 (B)/(A) of from 1.3 to 3.0, and more preferably from 1.5 to 2.5; the flow properties (B)/(A) being expressed using area (A) at the initial stage and area (B) after heating and pressing when, using glass sheets of 0.7 mm thick and 15 mm x 15 mm, a wiring-connecting material of 35  $\mu$ m thick and 5 mm x 5 mm  
 25 is held between the glass sheets and heated and pressured at 160°C and 2 MPa for 10 seconds. As long as the value is 1.3

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or more, the wiring-connecting material has sufficient flow properties and can achieve good connection. Also, as long as it is 3.0 or less, the wiring-connecting material is less likely to cause air bubbles to have a good reliability.

5           In addition, it is preferable for the wiring-connecting material of the present invention that its temperature at which exothermic reaction rises,  $T_a$ , is within the range of from 70°C to 110°C, having a peak temperature ( $T_p$ ) of  $T_a + 5$  to 30°C and an end temperature ( $T_e$ ) of 160°C or below, as measured with  
10   a differential scanning calorimeter (DSC) at a rate of temperature rise of 10°C/minute. Having such properties, the wiring-connecting material can achieve both low-temperature connecting performance and room-temperature storage stability.

          The wiring-connecting material of the present invention  
15   is also advantageous for reducing internal stress of the resin after connection and improving adhesive force, and also can achieve good conduction characteristics. Accordingly, it may preferably have a storage elastic modulus at 25°C after curing, of from 100 to 2,000 MPa, and more preferably from 300 to 1,500  
20   MPa.

#### J. Production of Wiring-connected Board

          The wiring-connecting material of the present invention is useful also as a filmlike adhesive for bonding IC chips to chip-mounting substrates and for bonding electric circuits to  
25   each other.

          More specifically, the use of the wiring-connecting

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material of the present invention enables production of a wiring-connected board in the following way: A first wiring member having a first connecting terminal and a second wiring member having a second connecting terminal are heated and pressed while disposing face to face the first connecting terminal and the second connecting terminal and providing the wiring-connecting material (filmlike adhesive) of the present invention between the first wiring member and the second wiring member, resulting in an electrical connection therewith.

10           The wiring member may include;  
            chips component parts such as semiconductor chips, resistor chips and capacitor chips;  
            printed-wiring substrates on which chips have been mounted and/or which have been resist-processed;  
15           TCPs (tape carrier packages) which comprise a TAB (tape-automated bonding) tape having chips mounted therewith and have been resist-processed; and  
            liquid-crystal display panels.

            It can be exemplified by;  
20           an insulating substrate comprised of silicon, gallium arsenide, glass, ceramics, a glass/thermosetting resin composite material (such as a glass/epoxy composite material) or a plastic material (such as a plastic film or a plastic sheet) of polyimide or the like on which substrate a conductive metal foil is formed  
25           via an adhesive to form a wiring which involves connecting terminals;

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an insulating substrate on which a conductive wiring has been formed by plating or vacuum deposition; and

a substrate coated with a material such as a plating catalyst and on which a conductive wiring has been formed.

5 As wiring members preferable for the connection carried out by the production process of the present invention, they may typically include TAB tapes, FPCs (flexible printed-circuit boards), PWBs (printed-wiring boards), ITO (indium tin oxide), and semiconductor chips having connecting pads.

10 Materials for the wiring member may be any of silicon or gallium arsenide for semiconductor chips, glass, ceramics, polyimide, a glass/thermosetting resin composite material (such as a glass/epoxy composite material) and plastics, without any particular limitations.

15 Where the surfaces of conductive connecting terminals coming into contact with the wiring-connecting material are formed of a transition metal such as copper or nickel, free radicals are generated as a result of its action of oxidation-reduction. Hence, radical polymerization may  
20 proceed when the wiring-connecting material is applied to the first connecting terminal for provisional adhesion and is left for a certain time, so that it may become difficult for the connecting material to flow. This may cause insufficient electrical connection at the time of final connection to the  
25 second connecting terminal registered. Accordingly, the surface of at least one of the connecting terminals may preferably

be constituted of at least one selected from gold, silver, a platinum group metal and tin. A plurality of metals may be used in combination, e.g., copper/nickel/gold, to make up a multi-layer configuration.

Use of such a plastic substrate allows the wiring-connected board to be thinner and lighter. In the production process of the present invention, the use of the wiring-connecting material of the present invention enables connection at a low temperature, and hence a plastic having a relatively low glass transition temperature or melting point can be used, making it possible to obtain economically advantageous wiring-connected boards.

10

## EXAMPLES

### Example 1

## 15

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tester method.

(2) Preparation of wiring-connecting material:

In solid content weight ratio, 40 g of the polyurethane resin A (as solid content) synthesized as described above, 39  
5 g of dimethyloltricyclodecane diacrylate, 1 g of phosphate type acrylate (available from KYOEISHA CHEMICAL Co., Ltd.; trade name: P2M), 20 g of phenoxy resin and 5 g of lauroyl peroxide (25 g as a methyl ethyl ketone solution) were mixed, and 3% by volume of conductive particles were further dispersed. The  
10 dispersion obtained was applied on a one-side surface-treated PET (polyethylene terephthalate) film of 80  $\mu\text{m}$  thick by means of a coater, followed by hot-air drying at 70°C for 10 minutes to obtain a wiring-connecting material having an adhesive layer of 35  $\mu\text{m}$  thick.

15 Here, as the radical-polymerizable substance, dimethyloltricyclodecane diacrylate was used. As the film-forming material, phenoxy resin (available from Union Carbide; trade name: "PKHC"; weight-average molecular weight: 45,000) was used. As the curing agent capable of generating  
20 a free radical upon heating, a 20% by weight methyl ethyl ketone solution of lauroyl peroxide [weight retention after open-leaving at room temperature (25°C) and normal pressure for 24 hours: 97%) was used. As the conductive particles, conductive particles having an average particle diameter of  
25 10  $\mu\text{m}$  were used which were produced by providing nickel layers of 0.2  $\mu\text{m}$  thick on the surfaces of particles whose cores were

(3) Connection of wiring:

As shown in Fig. 1 (a), using a laminated substrate  
11 provided with copper foil of 35  $\mu\text{m}$  thick, the copper foil  
was also patterned in a line width of 100  $\mu\text{m}$  and a pitch of  
200  $\mu\text{m}$  to form a circuit 12 and subjected to resist processing,  
and then gold was plated onto the copper foil surface to prepare  
a printed-wiring board (PWB) 10.

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registration [Fig. 1 (c)], where pressure 20 was applied with heating to effect interconnection to obtain a wiring-connected board 21 [Fig. 1 (d)].

## Example 2

### 5 (1) Synthesis of urethane acrylate:

400 parts by weight of polycaprolactone diol having an average molecular weight of 800, 131 parts by weight of 2-hydroxypropyl acrylate, 0.5 part by weight of dibutyltin dilaurate as a catalyst and 1.0 part by weight of hydroquinone  
10 monomethyl ether as a polymerization inhibitor were mixed while heating them to 50°C with stirring. Then, 222 parts by weight of isophorone diisocyanate was dropwise added, and further the temperature was raised to 80°C with stirring to carry out urethanation reaction. Making sure that the NCO conversion  
15 reached 99% or more, the reaction temperature was dropped to obtain urethane acrylate B.

### (2) Synthesis of polyimide resin:

#### An acid anhydride

2,2-bis[4-(3,4-dicarboxyphenoxy)phenyl]propane dianhydride  
20 (26.1 g) was dissolved in 120 g of cyclohexanone to obtain an acid dianhydride solution.

#### Diamines 2,2-bis[4-(4-aminophenoxy)phenyl]propane

(14.4 g) and  
1,3-bis(3-aminopropyl)-1,1,3,3-tetramethyldisiloxane (3.8 g)  
25 were also dissolved in 120 g of cyclohexanone to obtain a diamine solution.

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This diamine solution was dropwise added into a flask holding the acid dianhydride solution while controlling the temperature of the reaction system so as not to become higher than 50°C. After its addition was completed, the reaction mixture  
 5 was further stirred for 10 hours. Next, a Dean-Stark trap was attached to the system, 50 g of toluene was added and the temperature was raised to 120°C and kept it for 8 hours to effect imidization.

The solution obtained was cooled to room temperature,  
 10 and thereafter reprecipitated in methanol. The precipitate obtained was dried to obtain a polyimide resin having a weight-average molecular weight of 32,000. This was dissolved in tetrahydrofuran to form a 20% by weight polyimide solution C.

15 (3) Preparation of wiring-connecting material and production of wiring-connected board:

A wiring-connecting material was prepared, and a wiring-connected board was produced, in the same manner as in Example 1 except that 40 g of the polyurethane resin (as solid  
 20 content) synthesized in Example 1 was used as the polyurethane resin, 39 g of the urethane acrylate B synthesized in step (1) and 1 g of phosphate type acrylate were used in combination as the radical-polymerizable substance and 20 g of the polyimide resin C (as solid content) synthesized in step (2) was used  
 25 as the film-forming material.

Example 3

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As shown in Fig. 2, using a polyimide film with copper foil prepared in a double-layer configuration consisting of a polyimide film 22 and copper foil of 18  $\mu\text{m}$  thick, this copper foil was patterned in a line width of 100  $\mu\text{m}$  and a pitch of 200  $\mu\text{m}$  to form a circuit and a connecting terminal 23 and subjected to resist processing, and thereafter Au was plated onto the surface of the connecting terminal 23 to prepare a flexible printed-circuit board (FPC) 24. A wiring-connected board was obtained in the same manner as in Example 2 except that the TCP 19 was replaced with this FPC 24.

#### Example 4

Using a wiring-connecting material of 15  $\mu\text{m}$  thick, a wiring-connected board was obtained in the same manner as in Example 3 except that the printed-wiring board (PWB) 10 was replaced with a liquid-crystal display panel 27 comprising a glass substrate 25 and a connecting terminal and wiring 26 provided on the surface thereof using ITO.

#### Comparative Example 1

A wiring-connected board was obtained in the same manner as in Example 1 except for using a wiring-connecting material prepared by using a phenoxy resin (available from PKHC Union Carbide Corporation; trade name: PKHC; weight-average molecular weight: 45,000), a bisphenol-A epoxy resin (available from Yuka Shell Epoxy Kabushiki Kaisha; trade name: YL980) and an imidazole type microcapsular curing agent (available from Asahi Chemical Industry Co., Ltd.; trade name: 3941HP), setting the

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solid-content weight ratio of phenoxy resin/bisphenol-A epoxy resin/imidazole type microcapsular curing agent = 40/20/40, and mixing conductive particles therein in the same manner as in Example 1.

5                   Comparative Example 2

A wiring-connecting material was prepared, and a wiring-connected board was produced, in the same manner as in Example 1 except that the polyurethane resin A was replaced with a phenoxy resin (PKHC).

10                  Using the wiring-connecting materials and wiring-connected boards obtained in the foregoing Examples 1 to 4 and Comparative Examples 1 and 2, the adhesion, connection resistance, storage properties, insulating properties, polyurethane resin flow properties, wiring-connecting material  
15 flow properties, modulus of elasticity after curing, and DSC characteristics were measured or evaluated. Results obtained are shown in Table 1. Here, the measurement and evaluation were made in the following way.

(1) Measurement of adhesive force:

20                  The wiring-member connected article (wiring-connected board) obtained was separated in the direction of 90 degrees at a peel rate of 50 mm/minute to measure adhesive force. The adhesive force was measured immediately after the wiring-connected board was produced (initial stage) and after  
25 it was kept in a high-temperature high-humidity chamber of 85°C and 85%RH for 500 hours.

Using the wiring-connecting material obtained, a

The value of resistance between adjoining circuit lines was measured with a multi-meter at the initial stage and after keeping in a high-temperature high-humidity chamber of 85°C and 85%RH for 500 hours. The resistance value was shown as an average at 50 spots of resistance between the adjoining circuit lines.

The wiring-connecting material obtained was kept in a thermostatic chamber for 30 days, and a circuit was connected in the same manner as in the above (2) to evaluate storage stability.

Using the wiring-connecting material obtained, a

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μm, a pitch of 200 μm and a thickness of 18 μm were connected over a width of 2 mm by heating and pressing at 160°C and 3 MPa for 10 seconds. A voltage of 100 V was applied to the comb-shaped circuit of the resultant connected article, and  
 5 the value of insulation resistance after 500 hours of a 85°C and 85%RH high-temperature high-humidity test was measured.

(5) Measurement of flow point of polyurethane resin:

The temperature at which a cylinder begins to move when temperature is raised at a rate of 2°C/min under application  
 10 of a pressure of 3 MPa to make a sample flow out of a die of 1 mm in diameter was measured with a flow tester (manufactured by Shimadzu Corporation; trade name: CFT-100 Model).

(6) Evaluation of flow properties of wiring-connecting material:

15 Using a wiring-connecting material of 35 μm thick and 5 mm x 5 mm, this was held between glass sheets of 0.7 mm thick and 15 mm x 15 mm, and heated and pressed at 160°C and 2 MPa for 10 seconds. The value of flow properties (B)/(A) was determined using area (A) at the initial stage and area (B)  
 20 after heating and pressing, and regarded as an index of flow properties.

(7) Modulus of elasticity after curing:

The wiring-connecting material was immersed in 160°C oil for 1 minute to effect curing. Storage elastic modulus  
 25 of the cured film was measured (rate of temperature rise: 5°C/minute; 10 Hz) with a dynamic viscoelastometer to measure



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modulus of elasticity at 25°C.

(8) Measurement of DSC characteristics:

Using the wiring-connecting material obtained, the temperature at which exothermic reaction rises,  $T_a$ , the peak  
 5 temperature ( $T_p$ ) and the end temperature ( $T_e$ ) were determined when measured with a differential scanning calorimeter (DSC; manufactured by TA Instruments Japan Inc.; trade name: Model 910) at a rate of 10°C/minute.

Table 1

Items		Example 1	Example 2	Example 3	Example 4	Comparative Example 1	Comparative Example 2
Adhesive force (gf/cm)	Initial stage	1000	1000	800	1000	100	200
	85°C, 85%RH, 500h	800	900	600	900	peeled	peeled
Connection resistance (Ω)	Initial stage	2.2	2.3	2.1	2.1	90.6	2.1
	85°C, 85%RH, 500h	2.6	2.6	2.5	2.5	>500	2.6
Storage properties [connection resistance (Ω)]		2.2	2.4	2.3	2.2	120	2.3
Insulation resistance (Ω)	Initial stage	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <
	85°C, 85%RH, 500h	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>9</sup> <	1×10 <sup>6</sup>	1×10 <sup>9</sup> <
Flow properties		1.9	1.9	1.9	1.9	2.4	1.8
Modulus of elasticity (25°C)(MPa)		8001	600	600	600	1800	1400
Exothermic reaction (DSC) (°C)	Arising temperature (Ta)	89	92	92	92	98	86
	Peak temperature (Tp)	107	106	106	106	125	101
	End temperature (Te)	148	150	150	150	160	142

In all Examples, the adhesive force was approximately from 7.85 to 9.81 N/cm (800 to 1,000 gf/cm) as initial values, and was approximately from 5.88 to 8.83 N/cm (600 to 900 gf/cm) even after the test for resistance to moisture, showing a good adhesive force without any great decrease in bond strength. Comparative Example 1 showed insufficient curing reaction,

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and Comparative Example 2 showed a bond strength of about 1.96 N/cm (200 gf/cm) because the polyurethane resin was not used, resulting in a low adhesive force.

The wiring-connecting material obtained in Example 1  
5 had a low connection resistance at the initial stage and caused only a little increase in resistance after the high-temperature high-humidity test, showing a good connection reliability. The wiring-connecting materials of Examples 2, 3 and 4 and Comparative Example 2 also likewise achieved a good connection  
10 reliability. On the other hand, in Comparative Example 1, the curing reaction was so insufficient as to bring about a poor state of adhesion, resulting in a high connection resistance.

In Examples 1 to 4, results of connection were obtainable which were the same as those in a condition where the  
15 wiring-connecting materials were not treated in a 30°C thermostatic chamber for 30 days (i.e., at the initial stage).

In Examples 1 to 4, good insulating properties of  $1.0 \times 10^9 \Omega$  was also obtainable, and any lowering of insulating properties was observable.

20 With regard to the flow properties, both Example 1 and Example 2 showed a value of 1.9. Also, the modulus of elasticity of the wiring-connecting material of Example 1 at 25°C after curing was measured, and it was 800 MPa.

The wiring-connecting material of Example 1 also showed  
25 a curing reaction rise temperature of 89°C, a peak temperature of 107°C and an end temperature of 148°C. That of Example 2

showed a curing reaction rise temperature of 92°C, a peak temperature of 106°C and an end temperature of 150°C. This proves that they are curable at a lower temperature and, from the result of evaluation of storage properties, they have superior storage

5 properties.

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## 20

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1           1. A wiring-connecting material comprising from 2 to  
2   75 parts by weight of a polyurethane resin, from 30 to 60 parts  
3   by weight of a radical-polymerizable substance and from 0.1  
4   to 30 parts by weight of a curing agent generating a free radical  
5   upon heating.

1           3. The wiring-connecting material according to claim  
2   2, wherein said film-forming material is a polyimide resin.

1           5. The wiring-connecting material according to any one  
2 of claims 1 to 3, wherein said polyurethane resin has a flow  
3 point of from 40°C to 140°C as measured by the flow tester method.

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1           7. The wiring-connecting material according to any one  
2 of claims 1 to 6, wherein said radical-polymerizable substance  
3 is urethane acrylate.

1           8. A process for producing a wiring-connected board;  
2   the process comprising a connecting step of interconnecting  
3   wiring members each having a connecting terminal, which members  
4   are so interconnected that their connecting terminals are able  
5   to make conduction between them;

6           said connecting step comprising the step of heating  
7   the wiring-connecting material according to any one of claims  
8   1 to 7 while applying a pressure thereto via the wiring members;  
9   the wiring-connecting material being held between at least  
0   two wiring members so disposed that their sides having the  
1   connecting terminals face to each other.

9. The process for producing a wiring-connected board according to claim 8, wherein at least one of the connecting terminals has a surface formed of at least one metal selected from gold, silver and a platinum group metal.



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FIG. 1

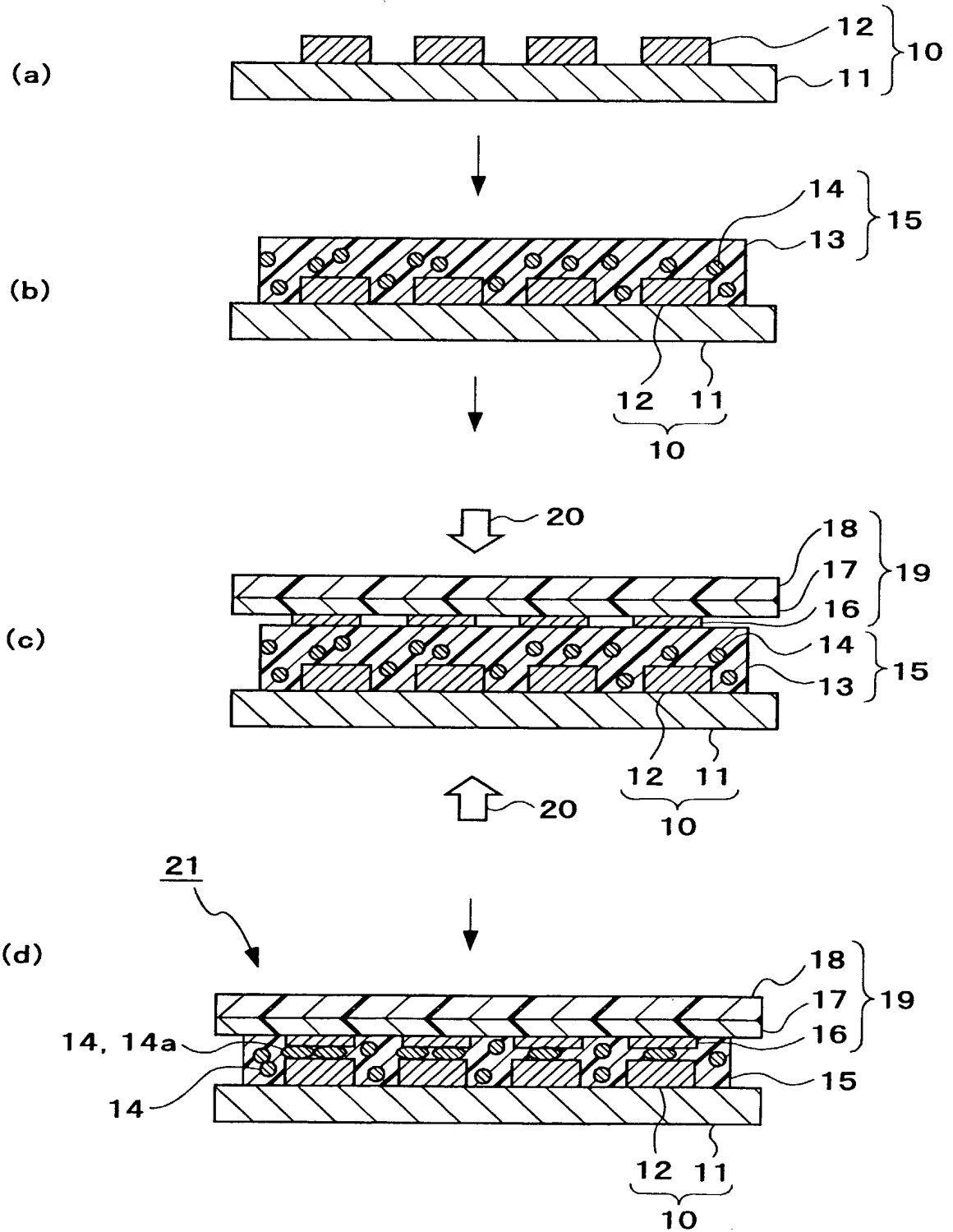




FIG.2

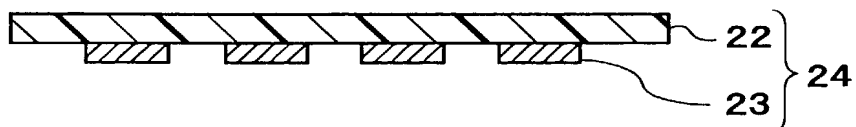
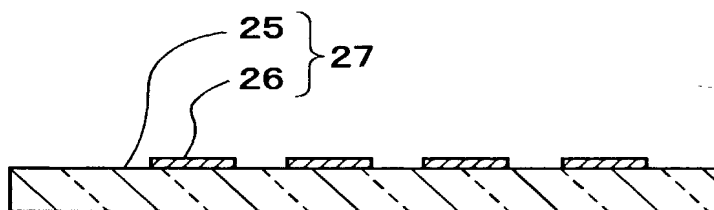


FIG.3



## Declaration and Power of Attorney for Patent Application

特許出願宣言書及び委任状

### Japanese Language Declaration

#### 日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

配線接続材料及びそれを用いた配線板製造方法

WIRING-CONNECTING MATERIAL AND

WIRING-CONNECTED BOARD PRODUCTION PROCESS

USING THE SAME

上記発明の明細書（下記の欄で印がついていない場合は、本書に添付）は、

the specification of which is attached hereto unless the following box is checked:

☒ 2000年8月25日に提出され、米国出願番号または特許協定条約国際出願番号を10/069053とし、  
（該当する場合）2002年5月21日に訂正されました。

☒ was filed on August 25, 2000  
as United States Application Number or PCT  
International Application Number 10/069053  
and was amended on  
May 21, 2002 (if applicable)

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.





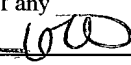
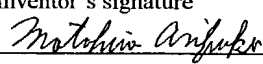

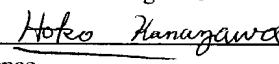
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Docket No. 566.41199X00

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